

“Limit” Model of CuO₂ Planes: Exact Results

Daniel C. Mattis

Department of Physics, University of Utah, Salt Lake City, Utah 84112

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We solve the simplest tight-binding model of electrons in a CuO₂ plane with Coulomb repulsion only on the copper ions, by a limiting procedure which retains *all* contributions $O(t^2/U)$ while projecting out the higher order terms. In addition to the ground state energy we identify a variety of quasiparticles; fermions with and without dispersion and localized spin-one triplets.

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Introduction and summary of results.—We solve the simplest possible model of a copper oxide (CuO₂) plane [1], in which the dynamics of the electrons is governed by a tight-binding Hamiltonian which is invariant under particle-hole conjugation

$$H = -t \sum_{i,\sigma} (c_{i,\sigma}^\dagger a_{i+\delta,\sigma} + \text{H.c.}) + H_2, \quad (1)$$

where i stands for the position of any of N copper ions on a square (sq) lattice $R_i = (n, m)$; $\delta = (\pm \frac{1}{2}, 0)$ or $(0, \pm \frac{1}{2})$ locates the four nearest ligand oxygens, and σ labels the spin. Here,

$$H_2 = U \sum_i [2(n_{i,\uparrow} - \frac{1}{2})(n_{i,\downarrow} - \frac{1}{2}) + \frac{1}{2}] \quad (2)$$

is the two-body interaction and $n_{i,\sigma} = c_{i,\sigma}^\dagger c_{i,\sigma}$ the occupation number operator of electrons on the copper ion.

The antiferromagnetic “mother phase” of high-temperature superconductors corresponds to occupation of each ligand p bond by two electrons (O^{2-}) and of each copper d orbital by one electron (Cu^{2+}) for a total of $n = 5$ per cell. Superconductivity is a feature of $n \approx 4.9$; however, this Letter does not directly concern superconductivity. Rather it presents a way to identify and classify the underlying quasiparticle eigenstates for $n \geq 4$ [2]. Because H_2 is a two-body interaction this normally poses an insoluble many-body problem. However, we have found a limiting procedure, specified below, whereby it is possible to obtain dynamical information for the low-lying states in dimensionless form.

The calculation proceeds in stages. First Fourier transform the c_i and $a_{i+\delta}$ operators. Because the CuO₂ lattice is a “decorated” sq lattice, a dispersionless band physically located on the oxygen ions (here denoted the “ β ” band) peels off. With Fermi level $\mu > 0$ the β band is fully occupied. We transform the surviving oxygen band (denoted the “ α ” band) and the copper band (the “ c ” band) to the Wannier representation. Now all operators are rooted on a common set of sq lattice sites R_i .

A trial ground state of singlet pairs centered about each lattice site yields a variational energy per cell $E/N = -29.374\,052 \dots t^*$, almost twice as low as for an equal number of particles in Bloch states in the Hartree-

Fock approximation [3]. We next sum all intersite contributions to the ground-state energy which are of $O(t^2/2U)$, proceeding to the limit $t \propto \sqrt{U} \rightarrow \infty$ while holding $t^* \equiv t^2/2U$ constant. We call this “the limit,” hence the title of this Letter. The exact ground-state energy for $n = 4$ is $E_0/N = -30.030\,539 \dots t^*$, close to the theoretical lower bound of $-32t^*$ [4]. All higher-order corrections vanish in the limit [5].

Added electrons or “holes” (i.e., some $n_i = 4 \pm 1$) form Kramers spin doublets ($S = \frac{1}{2}$) and are nominally [6] fermions carrying charge and crystal momentum $\hbar\mathbf{k}$ in identical [2] Bloch bands of width $W = 32.518\,700 \dots t^*$. A “hard-core” repulsion prevents more than one excitation from occupying a given site; other than that the fermions do not interact. The logarithmic van Hove singularity (vHS) in the one-particle density of states $\rho(\epsilon)$ is at $|\epsilon| = 22.994\,194 \dots t^*$, where the Fermi level would lie for $n = 5$ if there were no hard-core repulsion [7]. At low energies the dispersion in this “gapless semiconductor” is $|\epsilon| \propto |k|$, so that for $n \approx 4$ ($\mu \rightarrow 0$), $\rho(\mu) \rightarrow 0$ as shown in Fig. 1.

Additionally, above the vHS one finds localized spin $S = 1$, $n_i = 4$, states at energy $\epsilon = 29.374\,052 \dots t^*$. Once they are present they interact with the singlets and doublets and with each other [4] in a complicated

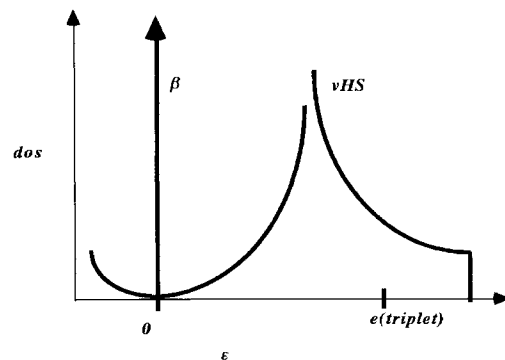


FIG. 1. One-particle density of states vs energy. Shown schematically are the electron ($n_i = 5$) and hole ($n_i = 3$) bands originating at $\epsilon = 0$, the van Hove singularity (vHS) and the energy of a triplet $S = 1$ excitation. The β band at zero energy accommodates up to $2N$ noninteracting electrons.

manner. Other eigenstates lie much higher, in the vicinity of $\varepsilon = U \rightarrow \infty$. The low-lying spectrum near $n = 4$ is schematized in the one-particle density of states (DOS) of Fig. 1).

In the simple model treated here, there are found no additional interactions among the quasiparticles besides the hard core, regardless of how many particles or excitations we introduce. The low-lying states of the Hamiltonian all have energies proportional to t^* . If we adopt this as the unit of energy our model is entirely devoid of adjustable parameters, except for temperature and Fermi level μ , the position of which determines the electron concentration.

Why is this model exactly soluble?—It should be emphasized that these exact results are contingent on several factors, including the particular form of Eq. (2) which is different from the various choices for this interaction adopted in the literature [1]. Any other form of the two-body interactions leads to a more complicated $n = 4$ ground state, including intractable “vacuum fluctuations.” In addition to the limiting procedure which projects out vast numbers of complicated diagrams and permits exact sums over intermediate states by use of the completeness theorem, there are several other aspects of our model which, on their face, are innocuous but which help attain closed-form solutions, viz., the assumption that $a_{i,\sigma}$, $b_{i,\sigma}$, and $c_{i,\sigma}$ are a complete set of *anticommuting* operators [8], i.e., that they (or their conjugate operators) destroy (or create) particles in *mutually orthogonal* localized orbitals, plus neglect of such other interactions as U_{pd} , $t_{pp'}$, and U_{pp} , $U_{pp'}$, etc. [9].

The calculations.—We now sketch the procedure. Fourier transformation of the operators in (1) yields

$$H = -t \sum_{k,\sigma} \omega(k) (c_{k,\sigma}^\dagger \alpha_{k,\sigma} + \text{H.c.}) + H_2, \quad (3)$$

where

$$\omega(k) = 2\sqrt{\cos^2 \frac{1}{2} k_x + \cos^2 \frac{1}{2} k_y}, \quad (4)$$

and

$$\alpha_{k,\sigma} = \frac{a_{k,\sigma} \cos \frac{1}{2} k_x + b_{k,\sigma} \cos \frac{1}{2} k_y}{\sqrt{\cos^2 \frac{1}{2} k_x + \cos^2 \frac{1}{2} k_y}}, \quad (5)$$

in which $a_{k,\sigma}$ is the Fourier transform of $a_{i+\delta,\sigma}$ operators on horizontal bonds and $b_{k,\sigma}$ of similar operators living on vertical bonds. A second linear combination of horizontal and vertical bonds, orthogonal to the α 's, is

$$\beta_{k,\sigma} = \frac{a_{k,\sigma} \cos \frac{1}{2} k_y - b_{k,\sigma} \cos \frac{1}{2} k_x}{\sqrt{\cos^2 \frac{1}{2} k_x + \cos^2 \frac{1}{2} k_y}}.$$

Together with its Hermitian conjugate $\beta_{k,\sigma}^\dagger$, the latter totally disappears from, and commutes with, the Hamiltonian. Consequently, the β band contains $2N$ localized (dispersionless) eigenstates for $SU(2)$ electrons at precisely $\varepsilon = 0$ with which to accommodate up to $2N$ electrons.

The corresponding *Wannier operators* include $c_{i,\sigma}$, the original copper orbital operator introduced in Eq. (1), and

$\alpha_{j,\sigma} = 1/\sqrt{N} \sum_{k \in \text{BZ}} e^{ik\mathbf{R}_j} \alpha_{k,\sigma}$, a linear combination of oxygen orbitals from a number of shells centered on the j th site. In its new representation the Hamiltonian takes the form

$$H = -t \sum_{i,j,\sigma} T(R_{ij}) (c_{i,\sigma}^\dagger \alpha_{j,\sigma} + \text{H.c.}) + H_2, \quad (6)$$

with $T(\mathbf{R})$ the lattice Fourier transform of $\omega(\mathbf{k})$:

$$T(\mathbf{R}) = \sum_{\mathbf{k} \in \text{BZ}} e^{ik\mathbf{R}} \omega(\mathbf{k}). \quad (7)$$

$T(\mathbf{0}) = 1.916\,182\,797$, $T(\pm 1, 0) = T(0, \pm 1) = 0.280\,185\,911$, $T(\pm 1, \pm 1) = -0.0470$, etc. The T 's drop off slowly with distance but there is, in fact, no need to discard any of them; the identity $\sum_{\text{all } \mathbf{R}} T^2(\mathbf{R}) = 4$ is used to sum all their contributions.

One starts with the terms in $T(\mathbf{0})$ and views intersite connections as perturbations. Decompose H as follows: $H = \sum_i H_i + H'$, including in each H_i all the terms relevant to a single site

$$H_i = -t \sum_{\sigma} T(\mathbf{0}) (c_{i,\sigma}^\dagger \alpha_{i,\sigma} + \text{H.c.}) + 2U(n_{i,\uparrow} - \frac{1}{2}) \times (n_{i,\downarrow} - \frac{1}{2}) + \frac{U}{2}, \quad (8)$$

while H' connects distinct pairs of sites (i, j) ,

$$H' = -t \sum_{i \neq j, \sigma} T(R_{ij}) (c_{i,\sigma}^\dagger \alpha_{j,\sigma} + \text{H.c.}). \quad (9)$$

So far, there have been no approximations, nor has the limiting procedure been invoked.

Of the 16 eigenstates of H_i (or 64 including the β 's) 8 (or 32) have energy $O(t^*)$ and the remaining 8 (or 32) energy $O(U)$. We list the low-lying states below. Integers indicating the occupancy (i.e., the charge) on the i th site will include two electrons assumed to be present in the passive β band; the second label, if any, indicates the spin $\sigma = \uparrow$ or \downarrow ($\pm \hbar/2$) in the usual notation, the last label, the site.

Low-lying eigenstates of each H_i .—At each site we can have one, and only one, of the following eight low-energy configurations [10].

(A) The $n = 3$ low-lying state is the Kramers doublet

$$|3_\sigma, i\rangle = \frac{c_{i,\sigma}^\dagger + p_3 \alpha_{i,\sigma}^\dagger}{\sqrt{1 + p_3^2}} |0\rangle,$$

where $p_3 = tT(\mathbf{0})/(U - e_3) \approx tT(\mathbf{0})/U$. Its energy is

$$e_3 = \frac{U}{2} - \sqrt{\left(\frac{U}{2}\right)^2 + [tT(\mathbf{0})]^2} \approx -\frac{[tT(\mathbf{0})]^2}{U} = -2T^2(\mathbf{0})t^*.$$

(B) The lowest energy belongs to $n = 4$,

$$|4, i\rangle = \frac{c_{i,\uparrow}^\dagger \alpha_{i,\downarrow}^\dagger + \alpha_{i,\uparrow}^\dagger c_{i,\downarrow}^\dagger + p_4 [\alpha_{i,\uparrow}^\dagger \alpha_{i,\downarrow}^\dagger + c_{i,\uparrow}^\dagger c_{i,\downarrow}^\dagger]}{\sqrt{2(1 + p_4^2)}} |0\rangle,$$

where $p_4 = 2tT(0)/(U - e_4) \approx 2tT(0)/U$ and

$$e_4 = \frac{U}{2} - \sqrt{\left(\frac{U}{2}\right)^2 + 4[tT(0)]^2} \approx -8T^2(0)t^*.$$

(C) The $n = 5$ doublet is

$$|5_\sigma, i\rangle = \frac{c_{i,\sigma}^\dagger \alpha_{i,-\sigma}^\dagger \alpha_{i,\sigma}^\dagger + p_5 c_{i,\sigma}^\dagger c_{i,-\sigma}^\dagger \alpha_{i,\sigma}^\dagger}{\sqrt{1 + p_5^2}} |0\rangle,$$

where $p_5 = p_3$ and $e_5 = e_3$. (D) Finally, the $n = 4$ zero energy $e_{S=1} = 0$ triplet states are

$$\begin{aligned} |\uparrow\uparrow, i\rangle &= c_{i,\uparrow}^\dagger \alpha_{i,\uparrow}^\dagger |0\rangle, & |\uparrow\downarrow, i\rangle &= \frac{c_{i,\uparrow}^\dagger \alpha_{i,\downarrow}^\dagger - \alpha_{i,\uparrow}^\dagger c_{i,\downarrow}^\dagger}{\sqrt{2}} |0\rangle, \\ |\downarrow\downarrow, i\rangle &= c_{i,\downarrow}^\dagger \alpha_{i,\downarrow}^\dagger |0\rangle. \end{aligned}$$

Intersite contributions and ground-state energy.—Even after omitting the β 's there remain (in principle) some 32 896 independent matrix elements of H' connecting *each and every pair of sites*. In the simplified model treated here, these boil down to just a few terms which can be handled without approximation.

The $|4, i\rangle$'s have by far the lowest energy and are prime candidates for the ground state, which for the sake of argument we shall call the “vacuum.” One can add particles or holes to this vacuum. It is, however, necessary to correct the vacuum energy for off-site correlations arising from H' , such as the bond between two sites i, j :

$$\Delta e_{ij} = -t^2 T^2(R_{ij}) \sum_\gamma |\langle \Phi_\gamma | \Omega_{ij} | 4, i \rangle \otimes | 4, j \rangle|^2 / (E_\gamma - E_0), \quad (10a)$$

in which $\Omega_{ij} = \sum_\sigma (c_{i,\sigma}^\dagger \alpha_{j,\sigma} + c_{j,\sigma}^\dagger \alpha_{i,\sigma}) + \text{H.c.}$ Upon calculating (10a) one finds that in leading order Ω_{ij} connects the ground state only to those excited states $\langle \Phi_\gamma |$ which have energy $O(U)$, and that the sum over such states can be evaluated in closed form.

That is the happy consequence of having chosen H_2 in the symmetric form of Eq. (2) [1]. Any other choice would have caused $\langle 4, i | \otimes \langle 4, j |$ to mix with the pair of low-lying excited states $\langle 3_\sigma, i | \otimes \langle 5_{-\sigma}, j |$, similar to “excitons.” $M \equiv \langle 4, i | \otimes \langle 4, j | \Omega_{ij} | 3_\sigma, i \rangle \otimes | 5_{-\sigma}, j \rangle$ just vanishes in our model by virtue of an internal symmetry. When M is not zero it is $O(t/U)$, hence, in general, the vertex for the transition $\langle 4, i | \otimes \langle 4, j | \Leftrightarrow \langle 3_\sigma, i | \otimes \langle 5_{-\sigma}, j |$ is $O(t^*)$, causing vacuum fluctuations which can neither be neglected nor calculated in closed form [11]. But even in such a case, where one is unable to write down the many-body $n = 4$ ground state in closed form, in the limit the energy still turns out proportional to t^* .

Because the energy denominator in (10a), $E_\gamma - E_0 = U$, is constant the “completeness theorem” serves to efficiently sum the numerator over *all* excited states.

$$\begin{aligned} &\sum_\gamma |\langle \Phi_\gamma | \Omega_{ij} | 4, i \rangle \otimes | 4, j \rangle|^2 / (E_\gamma - E_0) \\ &= \langle 4, i | \otimes \langle 4, j | \Omega_{ij}^2 | 4, i \rangle \otimes | 4, j \rangle / U = 2/U. \end{aligned} \quad (10b)$$

Combining (a) and (b) we obtain the bond energy

$$\Delta e_{ij} = -4t^* T^2(R_{ij}). \quad (10c)$$

Each is shared by two sites, thus the energy per site is just half, $\Delta e_{ij}/2$. Summation over all bonds at $R_{ij} \neq 0$ yields the complete ground-state *off-site* correlation energy

$$\Delta E = -2Nt^* \sum_{R \neq 0} T^2(R) = -2Nt^*[4 - T^2(0)], \quad (11)$$

using the sum rule. Combining this with the on-site energy, one finds the total ground-state energy to be exactly $E_0 = Ne_4 + \Delta E = -N(30.030539 \dots t^*)$.

If we replace the ground-state $|4, i\rangle$ singlet, together with all its bonds to other sites, by a Kramers doublet $|5_\sigma, i\rangle$, the effective “potential” energy cost is $V_i = (e_5 - e_4 + \text{the shift in total off-site bond energies})$. Once again as a consequence of the simple model assumptions, all off-site bond energies remain precisely unchanged and $V_i = e_5 - e_4 = 6T^2(0)t^*$ from (C) and (B). The (i, j) bond energy connecting two Kramers sites $[|5_\sigma, i\rangle \text{ replacing } |4, i\rangle \text{ and } |5_{\sigma'}, j\rangle \text{ replacing } |4, j\rangle]$ is also precisely $\Delta e_{ij} = -4t^* T^2(R_{ij})$ for all four choices of σ and σ' . Again this is no different from the vacuum bond energy, so that, aside from the hard core repulsion, the effective potential energy of interaction for two Kramers doublet states is seen to be $V(\mathbf{R}_{i,j}) \equiv 0$ in our model.

Quasiparticles, their motion, and their interactions.—Aside from the zero-range hard core we saw that there are no matrix elements which yield a finite two-body potential energy $V(\mathbf{R}_{i,j})$ [12], yet there are some in H' which help lift the translational degeneracy. Configuration $|4, i\rangle \otimes |5_\sigma, j\rangle$ is degenerate with $|5_\sigma, i\rangle \otimes |4, j\rangle$, in which charge is transported. We calculate the corresponding matrix element and, after lengthy algebra, find it to be precisely $+6T(R_{ij})T(0)t^*$. To keep track of an ever-increasing number of excited configurations and their motion it is helpful to introduce “quasiparticle” operators which operate on the vacuum, i.e., which create configurations of the type $|5_\sigma, i\rangle$ out of the $|4, i\rangle$ ground-state configurations. Denoting them $d_{5,\sigma}^\dagger(R_i)$, we can now write their Hamiltonian

$$H_d = 6t^* T(0) \sum_{ij} T(R_{ij}) d_{5,\sigma}^\dagger(R_i) d_{5,\sigma}(R_j). \quad (12a)$$

Included at $R_{ij} = 0$ is the energy V_i required to create a doublet in the first instance. In the Bloch representation, inversion of Eq. (7) and substitution into (12a) yields

$$\begin{aligned} H_d &= 6t^* T(0) \sum_{\mathbf{k} \in \text{BZ}} \omega(\mathbf{k}) d_{\mathbf{k},\sigma}^\dagger d_{\mathbf{k},\sigma} \\ &= 6t^* T(0) \sum_{\mathbf{k} \in \text{BZ}} \omega(\mathbf{k}) \hat{n}_{\mathbf{k},\sigma}; \end{aligned} \quad (12b)$$

$\hat{n}_{\mathbf{k},\sigma} = d_{\mathbf{k},\sigma}^\dagger d_{\mathbf{k},\sigma}$ is the occupation-number operator of a Bloch state in the “ d band.” The lowest Bloch energy $\varepsilon(k) = 6t^* T(0)\omega(k)$ is at (π, π) . Dispersion near the band minimum is thus a *linear* (rather than the usual quadratic) function of momentum. The DOS is $\rho(\varepsilon) \propto \varepsilon$, as in

Fig. 1. The vHS is at $\omega = 2$, i.e., at $\varepsilon = 12t^*T(0) = 22.994\,194\dots t^*$. As electrons are introduced, the energy changes to $\varepsilon(k) - \mu$. Here $\mu > 0$ and increases monotonically with the number of added electrons. Equations (12) supplemented by the *no double-occupancy* rule provide a genuine many-body Hamiltonian, essentially a t - J model (with $J = 0$) for the d quasiparticles. For a derivation of $J \neq 0$, see Ref. [9].

The band of hole quasiparticles which can be constructed out of the $|3_\sigma, i\rangle$ states is the mirror image of this d band when the average cell occupancy is 4. But as more electrons are added, the energy of the hole band increases as follows: $\varepsilon(k) + \mu$.

Triplets carry zero charge relative to the ground-state $|4, i\rangle$'s, hence H' cannot transport them in a background of $|4, i\rangle$'s and their energy is not changed by $\pm\mu$. The energy to create one triplet is $\varepsilon = 8T^2(0)t^* = 29.374\,052\dots t^*$, as quoted earlier. They are affected by the presence of d particles. With σ the spin operator of the fermion at R_i and S that of the spin-one entity at R_j the effective interaction is determined to be $H_{d\text{-triplet}} = t^*2T(0)T(R_{ij})[S \cdot \sigma + \frac{1}{2}]\mathbb{P}_{ij}$ where \mathbb{P}_{ij} is the permutation symbol which interchanges the two states. Triplets can annihilate against each other and against other excitations, thus they have a finite lifetime.

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- [1] In fact, a streamlined version of a model of CuO_2 was originally proposed by F.C. Zhang and T.M. Rice [Phys. Rev. B **37**, 37559 (1988)]. Such models have their ancestry in ideas proposed by J. Zaanen, G.A. Sawatzky, and J.W. Allen [Phys. Rev. Lett. **55**, 418 (1985)] to explain band gaps and anomalous electronic structures of transition-metal compounds, and for CuO_2 in observations by P.W. Anderson [Science **235**, 1196 (1987)] and V.J. Emery [Phys. Rev. Lett. **58**, 2794 (1987)]. Other relevant works include J.H. Jefferson, H. Eskes, and L.F. Feiner [Phys. Rev. B **45**, 7959 (1992)], who examined the limit of infinite U but finite, negative e_d , and L.F. Feiner [Phys. Rev. B **48**, 16857 (1993)]. It is generally believed that the tight-binding parameters (e.g., t_{pd} , etc.) of the copper

oxide plane can be obtained quantitatively by fitting to local density approximation (LDA) band structure calculations (even if band structure theory itself is placed in doubt). For an example, see J.M. Wheatley, Physica (Amsterdam) **215C**, 145 (1993).

- [2] By particle-hole symmetry, here, one only needs to know half of the states to infer the rest.
- [3] Our calculation for this yields $E_{\text{HF}}/N = -16t^*$ for $n = 4$.
- [4] D.C. Mattis, Mod. Phys. Lett. B **8**, 1387 (1994).
- [5] For example, the leading corrections are $O(t^4/U^3) = O(4t^{*2}/U) \rightarrow 0$. The limit model eliminates a large number of interactions and states without affecting essential symmetries of the problem, and is free of any arbitrary decisions as to which small terms to retain and which not.
- [6] When such particles are present in large numbers, their Fermi statistics might be transmuted [see R. Laughlin, Science **242**, 525 (1988) *inter alia*] but while this remains an interesting and vexing question, the resolution of the hard-core many-particle problem in 2D is not of our concern here.
- [7] This particular vHS is associated with "nesting," hence at $n = 5$ the metallic state is unstable against an energy gap even in the Hartree-Fock approximation.
- [8] The algebra of Fermi field operators which fail to anticommute can and does lead to a detailed explanation of superexchange. This rather interesting development in the context of the limit model will be reported in due course. [D.C. Mattis and J.M. Wheatley (unpublished).] The importance of the orbital's overlap integral in general, and the role of nonorthogonal atomic orbitals in a quantitative theory of superexchange, was already noted by P.W. Anderson [Phys. Rev. **115**, 2 (1959)] and is reprised in J.H. Jefferson, H. Eskes, and L.F. Feiner in the context of CuO_2 (Ref. [1]).
- [9] Although some parameters such as U_{pd} and $t_{pp'}$ can easily be incorporated into the theory, others such as U_{pp} once again render the model intractable.
- [10] Hence the hard-core condition on the composite quasiparticles.
- [11] This brings to mind a quote from Mattuck: "With the birth of general relativity around 1910 and quantum electrodynamics in 1930, the two- and one-body problems became insoluble. And within modern quantum field theory, the problem of zero bodies (vacuum) is insoluble. So if we are after exact solutions, no bodies at all is already too many!" [R.D. Mattuck, *A Guide to Feynman Diagrams in the Many-Body Problem* (McGraw-Hill, New York, 1976), p. 1].
- [12] Let alone an attractive $V(\mathbf{R}_{ij})$ with which to explain superconductivity.